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Short communication

Soil removal as a decontamination practice and radiocesium accumulation in tadpoles in rice paddies at Fukushima

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ABSTRACT

We investigated the biological accumulation of radiocesium in tadpoles [*Rana* (*Pelophylax*) *porosa porosa*] in rice paddies with and without decontamination practice at Fukushima. Radiocesium was accumulated in surface part of soils both in the control and decontaminated paddies one year after decontamination. Mean ^{134}Cs and ^{137}Cs concentrations in tadpoles in the control and decontaminated paddies were 3000 and 4500, and 600 and 890 Bq/kg dry weight, respectively. Radiocesium concentrations in surface soil (0–5 cm depth) and tadpoles in the decontaminated paddy were five times smaller than in the control paddy. These results suggest that decontamination practice can reduce radiocesium concentrations in both soil and tadpoles. However, at the decontaminated paddy, radiocesium concentrations in surface soils became 3.8 times greater one year after decontamination, which indicates that monitoring the subsequent movement of radiocesium in rice paddies and surrounding areas is essential for examining contamination propagation.

1. Introduction

Rice paddy is one of the dominant land-use types at Fukushima, and paddy areas were heavily contaminated by the Fukushima Daiichi nuclear power plant (FNPP) accident happened on March 11, 2011 (Yasunari et al., 2011; TEPCO, 2012). Among radionuclides emitted from FNPP, radiocesium (^{134}Cs and ^{137}Cs) are the dominant species, and expected to cause long-term contamination in the environment because of their long half-lives. Total ^{137}Cs deposition on the Japan islands was estimated more than 1 PBq (Yasunari et al., 2011), and local industries, including rice cropping and related agricultural activities, have suffered due to the accident. Rice paddies are also recognized as important landscape units for maintaining species diversity in a monsoon climate (MAFF, 2007). More than 5000 organisms inhabit rice paddies and related areas across the Japanese archipelago (Kiritani, 2010). Among them, 79% of frog species on the main islands of Japan specifically inhabit rice paddies or surrounding terrestrial and freshwater habitats, depending on their life stage (MAFF, 2010). Moreover, frog species can be keystone species because juvenile and adults are important food resources for birds, herptiles, and predatory insects (Blaustein and Kiesecker, 2002).

Radiocesium accumulation in tadpoles in rice paddies is an important component for understanding how the radiocesium potential transfers to higher trophic levels in paddy ecosystems. Tadpoles provide key links from lower to higher trophic positions because tadpoles are omnivores and are often preyed upon by other species (Bird et al., 1998; Alford and Richards, 1999). Based on experimental radionuclide additions in lake ecosystems in Canada, radionuclide accumulation tends to be higher in tadpoles than in slimy sculpin and leeches (Bird et al., 1998). Moreover, the concentrations of radionuclides in freshwater biota may depend on food, assimilation efficiency from food, metabolism, growth, and excretion (Thomann, 1981). Occurrence of the biological magnification of radionuclides depends on habitat conditions and food sources (Bird et al., 1998). Because organisms with high feeding rates tend to accumulate radiocesium (Håkanson and Andersson, 1992), tadpoles may have higher bioaccumulation. Although various studies have examined biological accumulation in lakes, rivers, and wetlands (Bird et al., 1998; Stark et al., 2004; Rowan, 2013), radionuclide deposition and biological accumulation in rice paddies had not been examined because we have not experienced radionuclide contamination in rice paddies in the past.

Various decontamination projects have been started as government campaigns after FNPP accident. Surface soil removal, hydrocleaning of buildings, and sediment removal along street

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gutter were proposed in the governmental guideline for decontamination in resident area (Ministry of the Environment, 2011). Also in agricultural fields including rice paddies, soil surface removal, tillage operation that buries contaminated soil surface, and removing suspended contaminated soil particles upon paddling in rice paddies were proposed (MAFF, 2011). They reported that the removal of surface soil reduced 75–97% of the radiocesium contamination immediately after decontamination programs were completed (MAFF, 2011), because deposited radionuclides on top of soil surface is generally adsorbed to fine soil particles, and rarely migrate to downward (Frissel and Pennders, 1983). Because particulate organic matter and clay minerals in paddy soil can adsorb to radiocesium, such contaminants may transfer radiocesium to organisms through food chain. However, the effect of decontamination on organisms inhabiting paddies is not known.

The objectives of this study were to examine radiocesium contamination in surface soil with and without decontamination practice and to investigate the biological accumulation of radiocesium in tadpoles in rice paddies. We then discuss the effects of decontamination practice on biological accumulation in paddy ecosystems. We hereby presented contaminations of soil and tadpoles after experimental decontamination in the rice paddies. Because we conducted this study during the earlier stages of the renaissance after the earthquake and FNPP accident, our data lacks replications for the experimental designs. Nevertheless, the data would be useful for understanding decontamination effects on paddy soils and tadpoles in the field based investigations.

2. Materials and methods

2.1. Study site and sampling

This study was conducted in Kawamata-cho, Fukushima Prefecture, approximately 40 km away from FNPP. Two adjacent rice paddies were selected as study sites (Fig. 1). Rice paddies are located typically flat areas surrounded by 10–30-cm banks that maintain approximately 10 cm of still water. In both paddies, water was supplied from the same drainage basin and drained independently via drainage channels installed in the lower portions of the paddies. On June 12, 2011, approximately 5–10 cm of surface soil was stripped from one rice paddy (decontaminated paddy) by heavy machinery for decontamination, whereas the other remained as an unmodified control paddy. After that, plowing in both paddies was conducted for experimental rice planting. Plowing was also conducted in spring in 2012 for the same purpose. On July 13, 2012, we collected tadpole samples of *Rana* (*Pelophylax*) *porosa porosa* that had hatched in 2012. The sampled individuals had not metamorphosed and did not have legs. We assumed that tadpole populations in two paddies were isolated, because outlet drainage channels were completely interrupted tadpole migration by weirs. Thus, the concentrations of radiocesium in tadpole samples were affected by the magnitude of contamination within each paddy. Four tadpole samples were collected using D-frame nets in each paddy; each sample contained approximately 100 individuals to provide sufficient mass for laboratory processing and analysis. The sampled tadpoles were preserved for half a day at ambient temperature to eliminate gut contents.

We collected five surface soil samples (0–5 cm) after the decontamination practice prior to plowing on June 12, 2011, and five soil core samples (ϕ 45 mm, vertically 20 cm deep from soil surface) on July 13, 2012 at 3-m intervals along the left side of levees approaching from water intake channels in each paddy. The soil core samples were frozen and separated into four positions (0–5, 5–10, 10–15, and 15–20 cm deep from the surface). Tadpole and soil samples were dried at 60 °C and 105 °C, respectively, for two days. Because soil samples did not contain large particles (>2 mm), tadpole and soil samples were directly pounded using an agate mortar and pestle, and preserved in 100-mL plastic containers prior to laboratory analysis. Water depth, the height of rice, grain size distribution, and soil organic matter content were similar between the paddies (Table 1).

2.2. Laboratory analysis

The radioactivity of ^{134}Cs and ^{137}Cs in the samples was determined by gamma-ray spectroscopy. Gamma-ray emissions at energies of 604.7 keV (^{134}Cs) and 661.6 keV (^{137}Cs) were measured using a high-purity germanium coaxial detector system (Ortec, GEM20-70) coupled to a multi-channel analyzer (Ortec, DSPEC jr 2.0). The energy and efficiency calibrations for this detector were performed using standard and blank (background) samples. For the analysis of radionuclide activity, each sample was measured for more than 1000 s. All activities were corrected for decay from the sampling date prior to statistical analyses.

We calculated concentration of radiocesium in tadpoles (Bq/kg wet weight) based on our dataset of Bq/kg dry weight and water contents of tadpoles (Burger and Snodgrass, 1998). Then, we estimated biota-sediment accumulation factor between tadpole and surface soil (0–5 cm) for ^{134}Cs and ^{137}Cs. The theoretical ratio of ^{134}Cs to ^{137}Cs in deposition on our sampling dates (June 12, 2011 and July 13, 2012) was calculated based on the half-lives of ^{134}Cs and ^{137}Cs (^{134}Cs: 2.1 y, ^{137}Cs: 30.1 y). We assumed that the amount of ^{134}Cs and ^{137}Cs fallout from FNPP was identical in this calculation (TEPCO, 2012).

2.3. Statistical analysis

Generalized linear mixed models (GLMMs) were constructed to evaluate decontamination, sampling date, and depth effects on radiocesium concentrations in soil ($n = 50$). Explanatory variables for radiocesium concentrations in soil were sampling site ($n = 2$), sampling date ($n = 2$), and depth ($n = 4$). Identifiers of individual surface soil samples in 2011, and soil core samples in 2012 (nos. 1–20) were included as random intercept in the models to give consideration to the expected variability in the spatial distribution of radiocesium. A Poisson error structure was used for the response variables together with a logit link function. Undetectable concentrations of radiocesium (n.d.) because of low values were regarded as 0 Bq/kg in these models.

Differences in radiocesium concentrations in tadpoles between the decontaminated and control rice paddies were tested using Student's *t*-test. Radiocesium concentrations in tadpoles were $\log_{10}(x + 1)$-transformed to normalize their distributions and standardize their variance structures prior to the tests. GLMMs were performed using the lme4 package in R 2.13.1 and Student's *t*-tests were performed using R 2.13.1 (R Development Core Team, 2011).

3. Results and discussion

The theoretical ratios of ^{134}Cs to ^{137}Cs in deposition on our sampling dates in 2011 and 2012 were 0.93 and 0.66, respectively. The ratios of soil surface (0–5 cm) collected in 2012 were from 0.57

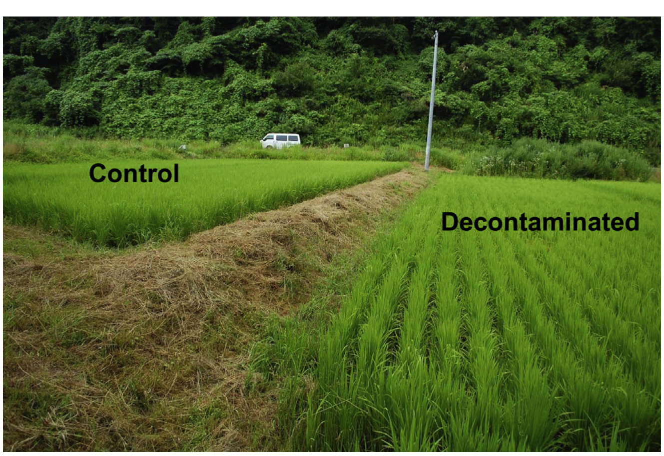

Fig. 1. Landscape of the decontaminated and control rice paddies in Kawamata-cho, Fukushima, Japan.

Table 1
Mean ± SE for water depth, height of rice, effective soil particle size (D_{50}), and soil organic matter content in the decontaminated and control rice paddies.

	n	Decontaminated	Control
Water depth (cm)	5	5.74 ± 0.20	4.88 ± 0.41
Height of rice (cm)	5	57.80 ± 0.80	57.80 ± 1.02
<I>D</I>$_{50}$ (μm)			
Depth: 0–5 cm	5	71.27 ± 5.52	72.57 ± 4.46
5–10 cm	5	94.26 ± 10.62	87.18 ± 6.22
10–15 cm	5	92.82 ± 9.92	96.18 ± 5.76
15–20 cm	5	110.42 ± 5.26	113.15 ± 10.53
Soil organic matter content (%)			
Depth: 0–5 cm	5	9.13 ± 0.31	9.15 ± 0.29
5–10 cm	5	8.54 ± 0.15	8.73 ± 0.72
10–15 cm	5	8.67 ± 0.18	8.75 ± 0.44
15–20 cm	5	8.01 ± 0.37	9.05 ± 0.77